Tail states in clean superconductors with magnetic impurities

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We analyse the behavior of the density of states in a singlet s-wave superconductor with weak magnetic impurities in the clean limit. By using the method of optimal fluctuation and treating the order parameter self-consistently we show that the density of states is finite everywhere in the superconducting gap, and that it varies as $\ln N(E) \propto -|E - \Delta_0|^{(7-d)/4}$ near the mean field gap edge Δ_0 in a d-dimensional superconductor. In contrast to most studied cases the optimal fluctuation is strongly anisotropic.

One of the most intriguing problems in modern condensed matter physics is the combined effect of correlations and disorder on the ground state and the electronic properties of solids. Interplay of superconductivity and impurity scattering is an example of a situation where such an effect is relevant experimentally, and where it has been studied for over forty years, beginning with the seminal papers by Anderson [1] and Abrikosov and Gor'kov (AG) [2]. Despite such a long history, however, the spectral properties even in the simplest case of a singlet s-wave superconductor continue to attract attention.

Since nonmagnetic impurities do not break the time-reversal symmetry, the "hard" gap, Δ_0, in the single particle excitation spectrum is unaffected by weak potential scattering [1]. At the same time scattering by magnetic impurities destroys the phase coherence in the superconducting state, and, consequently, leads to the suppression of the gap, and of the superconducting order parameter. AG analysed magnetic scattering using self-consistent Born approximation, and concluded that a hard gap in the energy spectrum survives up to a critical (average) concentration of weak magnetic impurities; it is followed, upon increasing impurity concentration, by a narrow region of gapless superconductivity, and then by destruction of the superconducting condensate.

After some early work [3], resurgence of interest in this problem started with the paper of Balatsky and Trugman [4], who argued that rare regions, where impurity concentration is sufficient to locally destroy superconductivity, yield a finite density of states (DOS) at the Fermi level. Their work was followed by other analyses [5]. Here we elucidate the nature of the subgap states, and provide a quantitative analysis of the energy profile of the DOS in s-wave superconductors with weak magnetic impurities.

In the AG theory the effect of disorder is controlled by a dimensionless parameter, $\Delta\tau_s$, where τ_s is the scattering time due to magnetic impurities, and Δ is the value of the superconducting order parameter. In particular, the single particle spectral gap is $\Delta_0 = \Delta(1 - (\Delta\tau_s)^{-2/3})^{3/2}$, indicating the onset of the gapless superconductivity at $\Delta\tau_s = 1$. In the regime studied here, $\Delta\tau_s \gg 1$, the AG theory predicts a gapped quasiparticle spectrum.

These results are obtained by carrying out a standard impurity averaging procedure. It is clear, however, that, among all the realizations of the impurity distribution, there exist regions where the resulting potential generates localized quasiparticle states at an arbitrary energy below the gap edge. Such localized states were extensively studied in doped semiconductors [6, 7]. For a particular energy, E, the most probable (albeit still rare) configuration of impurities that creates a state at E, and therefore contributes the most to the DOS, $N(E)$, is called the optimal fluctuation (OF) [6]. Such rare regions provide nonperturbative corrections to the DOS determined in the framework of self-consistent Born approximation.

We employ the OF method in a singlet s-wave superconductor with magnetic impurities. We also consider the self-consistent suppression of the superconducting order parameter; it is known that in d-wave superconductors it significantly affects the low-energy DOS [8]. We find that the density of states is finite everywhere in the superconducting gap. Just below the AG gap edge, the DOS is $N(E)/N_0 \propto \exp\left(-|E - \Delta_0|^{(7-d)/4}\right)$, where N_0 is the normal state DOS, and d is the number of spatial dimensions. In contrast to other known cases, the OF is anisotropic, with its transverse size much smaller than the longitudinal extent.

We consider a singlet s-wave superconductor. In the 4-space of the wave functions $\left(\psi_\uparrow^\star(\mathbf{r}), \psi_\downarrow^\star(\mathbf{r}), \psi_\uparrow(\mathbf{r}), \psi_\downarrow(\mathbf{r})\right)$, the mean field hamiltonian is

$$\widehat{H} = \widehat{\xi}\tau_3 + \Delta(\mathbf{r})\tau_1\sigma_2 + \widehat{U} \qquad (1)$$

Here $\widehat{\xi} = -\nabla^2/(2m) - \mu$ is the kinetic energy of a quasiparticle with respect to the Fermi level, μ, and τ_i and σ_i are the Pauli matrices in the particle-hole and the spin space respectively, so that $\tau_i\sigma_j$ is a 4×4 direct product.

Potential due to impurities, $\widehat{U}$, includes both potential and spin-flip scattering processes. When the potential scattering is dominant (motion of quasiparticles in the OF is diffusive) properties of the low-energy states were explored in Ref. [5]. However, there exists experimental

evidence that in some situations the magnetic scattering is dominant: upon increasing the impurity concentration the increase in residual resistivity ratio correlates with the suppression of the superconducting transition temperature [9]. Guided by this insight, we consider only the magnetic scattering, and expect our results to remain valid for as long as it is stronger than, or is of the order of, potential scattering. Hence we write $\widehat{U} = \mathbf{U}(\mathbf{r}) \cdot \mathbf{s}$, where $\mathbf{s}$ is the electron spin operator, $\mathbf{U}(\mathbf{r}) = \sum_i J\mathbf{S}_i\delta(\mathbf{r}-\mathbf{r}_i)$, J is the exchange constant, and $\mathbf{S}_i$ is the localized impurity spin at a site i.

The main physical difference between our analysis and that of Ref. [5] lies in this choice of the scattering potential. In the regime studied here the mean free path significantly exceeds the coherence length, and hence the motion of the quasiparticles in the optimal fluctuation is ballistic, leading to a substantially different physical picture of the tail states and the optimal fluctuation, and to a different energy dependence of the density of states.

The states with energy $E \lesssim \Delta_0$ exist in rare regions where the amplitude of the impurity potential differs significantly from its typical value. Therefore in determining $N(E)$ it is sufficient to consider only such configurations of the impurity potential, for which E is the lowest quantum mechanical energy level; fluctuations where E is the energy of a higher bound state are exponentially less probable [7]. Also, in essentially all the energy range below the gap the size of the optimal fluctuation is significantly greater than the distance between impurities, so that the exact impurity potential can be replaced by a smooth function, averaged over regions containing many impurities, but smaller than the characteristic size of the wave function in the optimal fluctuation [7].

Hence we consider an uncorrelated potential with a gaussian probability density

$$P[\mathbf{U}] \propto \exp\left(-\frac{1}{2U_0^2}\int d^d\mathbf{r}\mathbf{U}^2(\mathbf{r})\right), \qquad (2)$$

where $U_0^2 = n_{imp}J^2S(S+1)/3$ is related to the scattering time via $\tau_s^{-1} = 2\pi N_0 U_0^2$, and n_{imp} is the average impurity concentration. We ignore interactions between the magnetic impurities: it was shown in Ref. [10] that the RKKY interaction and glassy behavior of impurity spins modify the AG results very weakly. We also do not include quantum dynamics of the impurity spins, and therefore cannot account for the Kondo effect. This is justified either when the Kondo temperature of individual impurity sites is much smaller than the superconducting transition temperature, $T_K \ll T_c$ (and depletion of states at the Fermi level prevents screening of the local moment), or in the opposite limit, $T_K \gg T_c$, when the moments are quenched already in the normal state [11].

The density of localized states in the fluctuation region of the spectrum is then [7]

$$N(E) = \int \mathcal{D}\mathbf{U}P[\mathbf{U}]\delta(E - \mathcal{E}[\mathbf{U}]), \qquad (3)$$

where $\mathcal{E}[\mathbf{U}]$ is the lowest energy eigenstate in the realization $\mathbf{U}$ of the impurity potential. For rare configurations the integral is evaluated using saddle point approximation to give $\ln[N(E)/N_0] \approx -\mathcal{S}[\mathbf{U}_{opt}]$, where the optimal fluctuation is obtained by minimizing the functional

$$\mathcal{S}[\mathbf{U}] = \frac{1}{2U_0^2}\int d^d\mathbf{r}\mathbf{U}^2(\mathbf{r}) + \lambda\left(\mathcal{E}[\mathbf{U}] - E\right) \qquad (4)$$

with respect to both $\mathbf{U}$ and the Lagrange multiplier λ. The difficulty in minimizing the action is in the nonlinear nature of the equations: optimal potential $\mathbf{U}$ depends on the wave function of the particle in this potential.

The method of optimal fluctuation allows for a simple physical analysis. Consider first a semiconductor. In a potential well of depth U (all energies are measured from the band edge) and size L the energy of the localized state is of the order of $U + 1/(mL^2) = E$ ($\hbar = 1$). In the optimal fluctuation $E \sim U \sim L^{-2}$, so that the action for such fluctuation is $\mathcal{S}[U] \approx L^d U^2/U_0^2$, or $\ln[N(E)/N_0] \approx -|E|^{2-d/2}/U_0^2$. This is exactly the result obtained by Lifshits, and is confirmed by the solution of the nonlinear equations for minimization of action in Eq.(4) [6, 7].

The difference between a potential well in a doped semiconductor and in a superconductor is twofold. First, because of particle-hole mixing the hamiltonian Eq.(1) is a matrix in particle-hole and spin space. Second, we are concerned with quasiparticles close to the Fermi energy.

We assume that ferromagnetic fluctuation maximizes the effect of the impurity potential [12]. Consequently, we consider such a fluctuation, and choose the direction of the impurity spins along the y-axis, so that $\widehat{U} = U(\mathbf{r})\sigma_2$. Performing a spin rotation, $\sigma_2 \to \sigma_3$ in Eq.(1), we obtain a hamiltonian diagonal in the spin space,

$$\widehat{H}_{\pm} = \widehat{\xi}\tau_3 \pm \Delta_0\tau_1 \pm U(\mathbf{r}). \qquad (5)$$

It is therefore sufficient to consider only one spin orientation. Let us again consider the problem qualitatively, and concentrate first on the one dimensional case ignoring the suppression of the order parameter. We linearize the kinetic energy near the Fermi surface, $\widehat{\xi} \approx -iv_F(\partial/\partial x)$, so that typical kinetic energy in an OF of size L is $\xi \simeq v_F/L$. Then the energy of a quasiparticle in the optimal fluctuation (measured from the Fermi energy) is $E \simeq U + \sqrt{\Delta_0^2 + v_F^2/L^2}$. For the energies close to the superconducting gap, $(\Delta_0 - E)/\Delta_0 \ll 1$, the OF is large ($L \gg \xi_0 = v_F/\Delta_0$) and shallow ($|U|/\Delta_0 \ll 1$), so that $E - \Delta_0 \approx U + v_F^2/(\Delta_0 L^2)$. Introducing the dimensionless energy $\epsilon = E/\Delta_0$, we obtain, in analogy with the arguments above, $|U|/\Delta_0 \simeq \xi_0^2/L^2 \simeq 1 - \epsilon$. Notice that the size of the fluctuation is indeed $L \simeq \xi_0/\sqrt{1 - \epsilon} \gg \xi_0$. As a result, we find $\mathcal{S}[U] \approx LU^2/U_0^2 = \Delta_0^2\xi_0(1 - \epsilon)^{3/2}/U_0^2$. From the definition of U_0 it follows that

$$-\ln\frac{N(E)}{N_0} \approx \mathcal{S}[U_{opt}] \simeq (\Delta_0\tau_s)(1 - \epsilon)^{3/2}. \qquad (6)$$

The energy dependence in Eq.(6) is identical to the result of Lifshits in $d = 1$. This follows from the expansion in $\xi \ll \Delta_0$: even though $\xi \propto 1/L$, the expansion is in ξ^2 .

We now verify these estimates by a considering the energy of a particle in hamiltonian Eq.(5) for spin "up"

$$\mathcal{E}_+[U] = \langle \widehat{H}_+ \rangle = \langle \Psi | \widehat{\xi} \tau_3 + \Delta_0 \tau_1 + U | \Psi \rangle, \qquad (7)$$

where Ψ is the normalized wave function of the particle. Minimization of Eq.(4) with respect to U gives

$$U(x) = -\lambda U_0^2 \langle \Psi | \frac{\delta \widehat{H}_+}{\delta U} | \Psi \rangle, \qquad (8)$$

while minimization with respect to λ dictates $\widehat{H}_+ | \Psi \rangle = E | \Psi \rangle$. We first ignore the self-consistent suppression of the gap, which means $U(x) = -\lambda U_0^2 (\Psi^\star(x) \Psi(x))$, where $(\Psi^\star \Psi)$ denotes the scalar product in particle-hole space. Then the Schrödinger equation takes the form

$$\left[-i v_F \frac{\partial}{\partial x} \tau_3 + \Delta_0 \tau_1 - \lambda U_0^2 (\Psi^\star \Psi) \right] \Psi = E \Psi. \qquad (9)$$

This equation is solved by introducing the bilinear forms $\Psi^\star(x) \tau_i \Psi(x)$, and yields the optimal fluctuation

$$\frac{U(x)}{2\Delta_0} = -\frac{1 - \epsilon^2}{\epsilon + \cosh(2x\sqrt{1-\epsilon^2}/\xi_0)}, \qquad (10)$$

which corresponds to the value of the action

$$\mathcal{S}[U] = 8\pi(\Delta_0 \tau_s) \left[\sqrt{1-\epsilon^2} - \epsilon \arccos \epsilon \right]. \qquad (11)$$

We immediately notice that for $\epsilon \approx 1$ the length scale of the optimal fluctuation is $\xi_0/\sqrt{1-\epsilon^2}$, its depth is $U \sim \Delta_0(1-\epsilon^2)$, and the action $\mathcal{S}[U_{opt}] \simeq (8\pi/3)(\Delta_0 \tau_s)(1-\epsilon^2)^{3/2}$, in complete agreement with our estimates above.

We now show that the self-consistent suppression of the order parameter does not appreciably change our result. Self-consistency is achieved by including the variation of the gap into the variational derivative $\delta \widehat{H}_+ / \delta U$. We notice that, at $T = 0$, uniform U does not suppress superconductivity. Consequently, Δ depends on the gradient of the potential. For $1 - \epsilon \ll 1$, the potential varies smoothly, so that $dU/dx \ll \Delta_0/\xi_0$ can be accounted for perturbatively. The leading local correction to the gap is

$$\frac{\delta\Delta(x)}{\Delta_0} = -\frac{1}{6} \frac{\xi_0^2}{\Delta_0^2} \left(\frac{dU}{dx} \right)^2, \qquad (12)$$

and the correction to the action for $\epsilon \sim 1$ is $\delta\mathcal{S} \simeq (128\pi/315)(\Delta_0 \tau_s)(1-\epsilon^2)^{7/2}$. At lower energies the self-consistent $\Delta(x)$ has to be computed numerically. The results are presented in Figs. 1 and 2. It is clear that the suppression of the gap, even for the states with $E \ll \Delta_0$ is incomplete; since $|U| \leq 2\Delta_0$, the gap remains at a
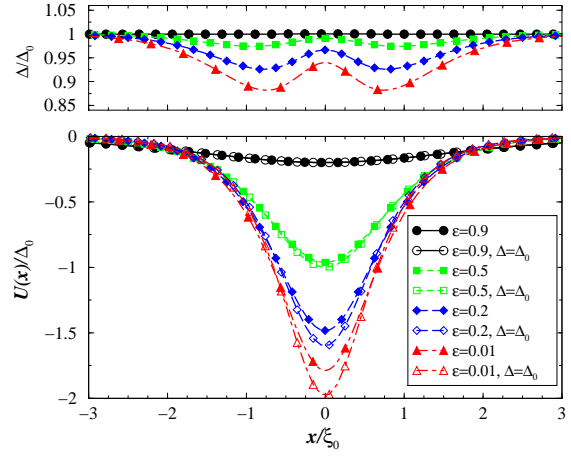

FIG. 1: Bottom panel: comparison of the optimal potential for $\Delta = \Delta_0$ from Eq.(10) with that obtained from the self-consistent numerical solution, for different values of the bound state energy $E = \varepsilon\Delta_0$. Top panel: the corresponding self-consistently determined profile of the order parameter. Maximum in $\Delta(x)$ at $x = 0$ corresponds to small gap suppression at an extremum of the potential, see Eq.(12).

significant fraction of Δ_0 throughout the OF. This justifies the expansion in the bare energy, ξ, in our qualitative analysis. Consequently, the optimal action computed self-consistently differs at most by 10% from that computed assuming a uniform gap, see Fig. 2.

Having demonstrated that the qualitative consideration are in excellent agreement with the full solution of the problem in $d = 1$, we discuss the multidimensional case. In a doped semiconductor the OF in any d is spherically symmetric [6, 7]. This results from the balance between lowering the particle energy in a large and deep fluctuation, and the probability cost of such an OF.

Since electrons in a superconductor move with the Fermi velocity, the wave function of the subgap state is concentrated along the quasiclassical trajectory, which is a chord in a potential of any shape. Consequently, there is little energy cost in reducing the size of the OF in the "transverse" direction, while the smaller volume makes such fluctuations more probable. As a result, the optimal fluctuation is anisotropic, and strongly elongated in one direction. Choosing this direction as the x-axis, we can write the wave function of the subgap state as $\Psi(x, \mathbf{y}) = \exp(ik_F x)\Phi(x, \mathbf{y})$, where $\mathbf{y}$ denotes the transverse $d-1$ coordinates, and Φ is a slowly varying function. Therefore the kinetic energy of the quasiparticle is

$$\widehat{\xi}\Psi \approx -e^{ik_F x} \left(i v_F \frac{\partial}{\partial x} + \frac{\nabla_y^2}{2m} \right) \Phi \sim \left(\frac{v_F}{L_x} + \frac{1}{mL_y^2} \right) \Psi. \qquad (13)$$

The transverse size of the fluctuation can therefore be reduced until the second term becomes comparable to the first, i.e. $L_y \simeq (\lambda_F L_x)^{1/2}$, where $\lambda_F \simeq k_F^{-1}$ is the
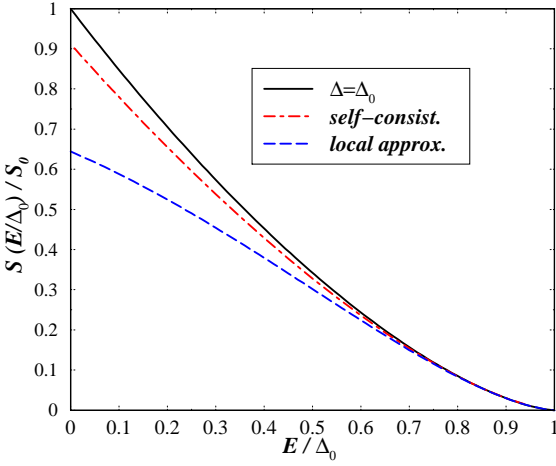
FIG. 2: Optimal action, normalized by $\mathcal{S}_0 = 8\pi(\Delta_0 \tau_s)$. Local approximation: $\Delta(x) = \Delta_0 + \delta\Delta(x)$, with $\delta\Delta(x)$ from Eq.(12).

Fermi wavelength. Consequently, we find $|U|/\Delta_0 \sim 1 - \epsilon$ and $L_x \sim \xi_0/\sqrt{1 - \epsilon}$, and

$$\mathcal{S}[U_{opt}] \simeq L_x L_y^{d-1} \frac{U^2}{U_0^2} \simeq (\Delta_0 \tau_s) \left(\frac{E_F}{\Delta_0}\right)^{\frac{d-1}{2}} (1 - \epsilon)^{\frac{7-d}{4}},$$
(14)

where E_F is the Fermi energy. Eq.(14) is the main result presented here. The action for the anisotropic fluctuation is smaller than that for an isotropic OF, by a factor of $(E_F/\Delta_0)^{(d-1)/2}(1-\epsilon)^{-(d-1)/4}$, so that the corresponding DOS is exponentially higher.

There are two limitations on the validity of the results obtained here. First, since the optimal fluctuation is a result of a saddle point approximation for the functional integral, Eq.(3), it is only valid when $\mathcal{S}[U_{opt}] \gg 1$, or

$$1 - \epsilon \gg (\Delta_0 \tau_s)^{\frac{4}{d-7}} \left(\frac{\Delta_0}{E_F}\right)^{\frac{2(d-1)}{7-d}}.$$
(15)

For $d = 1$ this condition becomes $1 - \epsilon \gg (\Delta_0 \tau_s)^{-2/3}$, while for $d = 3$ it does not depend on the gap amplitude, $1 - \epsilon \gg (k_F l)^{-1}$, where $l = v_F \tau_s$ is the mean free path. In $d = 1$ the region of validity is extended by almost an order of magnitude in comparison to this estimate as the action has a large numerical factor ≈ 24.

Second, when the characteristic size of the OF $L \geq l$, our assumption about the ballistic motion in the fluctuation is invalid, and a crossover to the diffusive regime studied in Ref. [5] occurs for $1 - \epsilon \leq (\Delta_0 \tau_s)^{-2}$ in any dimension. When $d = 1$ for $(\Delta_0 \tau_s) \geq 5$ the saddle point approximation becomes invalid before the diffusive regime is reached. For $d > 1$ and typical $\Delta_0/E_F \simeq 10^{-3}$, the OF method works up to the crossover. Taking $(\Delta_0 \tau_s) \sim 10$, we find that our results hold to within 1% of Δ_0, while the expansion in ξ is quantitatively valid for $\epsilon \geq 0.9$, and

qualitatively for $\epsilon \geq 0.75$, providing a significant window of applicability for our DOS.

Experimental verification of our results, and the underlying physical picture of scattering on randomly distributed impurities, requires averaging the tunneling conductance over regions containing many impurities, and is best done at energies just below Δ. We suggest averaging the tunneling spectra, obtained from Scanning Tunneling Spectroscopy, over several distinct areas of the sample, each containing a large number of impurities.

To summarize, we have analysed the density of subgap states in an s-wave superconductor with weak magnetic impurities using the method of the optimal fluctuation. We concentrated on the clean limit, $l \gg \xi_0$, when the motion of particles in the optimal potential is ballistic. We find that the optimal fluctuation in this case is strongly anisotropic, and that the density of states varies as a stretched exponential below the gap edge, with a power that depends on the dimension.

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